

Using granular Co-Al₂O₃ spacer for optimization of functional parameters of the FeMn/Fe₂₀Ni₈₀ magnetoresistive films

A N Gorkovenko, V N Lepalovskij, O A Adanakova and V O Vas'kovskiy

Department of Magnetism and Magnetic Nanomaterials, Ural Federal University,
Ekaterinburg 620002, Russia

E-mail: a.n.gorkovenko@urfu.ru

Abstract. In this paper we studied the possibility of tailoring the functional properties of the multilayer magnetoresistive medium with unidirectional anisotropy and the anisotropic magnetoresistance effect (AMR). Objects of the research were composite Co-Al₂O₃ films and Ta/Fe₂₀Ni₈₀/Fe₅₀Mn₅₀/Fe₂₀Ni₈₀/Co-Al₂O₃/Fe₂₀Ni₈₀/Ta multilayers structures obtained by magnetron sputtering and selectively subjected vacuum annealing. Structure, magnetic and magnetoresistive properties of the films in the temperature range 77÷440 K were investigated.

1. Introduction

Magnetic films containing exchange-coupled antiferromagnetic and ferromagnetic layers are widely used in magnetic field sensors based on the effect of giant magnetoresistance. However, in some cases, the value of the output signal is not as important as the functionality of the magnetic sensors, for example, odd conversion function [1]. For such purpose, it is more appropriate to use medium which has both the exchange bias and the anisotropic magnetoresistance (AMR). The main objective of this work was to determine the optimal structural characteristics of this medium in the classical ferro-antiferromagnetic FeMn/Fe₂₀Ni₈₀ system. As was shown earlier [2] its practical potential is realized in a SiO₂/Ta(5)/Fe₂₀Ni₈₀(5)/Fe₅₀Mn₅₀(20)/Fe₂₀Ni₈₀(45)/Ta(5) multilayer. The presence of two layers of permalloy (seed layer with thickness of 5 nm and thicker layer is responsible for AMR) allows realization of the exchange bias and ensure the thermal stability of the exchange bias field H_e . At the same time within this structure variation of H_e is complicated which means it is difficult to regulate intentionally the sensitivity of the magnetic field sensors. Efficiency of the interlayer exchange coupling can be changed by introduction of ultra-thin non-magnetic spacer (for example, Al₂O₃) between the interacting layers. However, direct separation of Fe₅₀Mn₅₀ and Fe₂₀Ni₈₀(45) layers, even with a very thin spacer (thickness L is no more than 0.2 nm), leads to the absence of the magnetic exchange bias. Therefore, the spacer introduced into the permalloy layer, which is responsible for AMR, divides it into two unequal parts. The smaller one with thickness of 5 nm is directly adjacent and exchange coupled to the antiferromagnetic layer and larger one with thickness of 40 nm is providing AMR. Furthermore, in order to impart additional features of the spacer for variation of the interlayer interaction the spacer was enriched with Co which is localized in Co_x(Al₂O₃)_{100-x} with $x < 50\%$ in form of nanoscale granules [3]. Thus, the films which were investigated in this work had the following general structural formula SiO₂/Ta(5)/Fe₂₀Ni₈₀(5)/Fe₅₀Mn₅₀/Fe₂₀Ni₈₀(5)/Co-Al₂O₃(L)/Fe₂₀Ni₈₀(40)/Ta(5).



2. Samples and techniques

Composite Co-Al₂O₃ films with thickness of 100 nm and multilayers with variable thickness of Co-Al₂O₃ spacer were prepared by magnetron sputtering (Orion sputtering system, AJA International, Inc.) onto Corning glass substrates. The composite layers were formed by co-sputtering of Co and Al₂O₃ targets. Other layers were deposited using homogeneous targets of appropriate compositions. Films were deposited at presence of the uniform magnetic field which was used to ensure unidirectional anisotropy. A series of samples was annealed in vacuum at 350 °C for one hour in order to increase AMR effect. Structure and elemental composition of the films were investigated by Carl Zeiss AURIGA CrossBeam scanning electron microscope and high-resolution 30 CM Super Twin transmission electron microscope. Magnetic properties and magnetoresistance of multilayers were determined using the LakeShore 7407VSM measurement system at temperatures ranging from 77 to 440 K.

3. Results and discussion

As was mentioned above in composites such as Co-Al₂O₃ cobalt can be in granular form. Moreover, the granules at room temperature due to the small granule size demonstrate superparamagnetic behavior. However, being a part of the ultra-thin spacer, which separate magnetically ordered layers, the granules can contact them, become ferromagnetically ordered and serve as a bridges for the implementation of the interlayer exchange coupling at a greater distance. In this context, important factors determining the efficiency of the exchange separation of the main layers are composition and thickness of the spacer.

In a preliminary experiment carried out on single-layer films, influence of the composition on the structure and chemical state of the Co_x(Al₂O₃)_{100-x} composite was evaluated. The composition was determined by the ratio of partial deposition rates of Co и Al₂O₃. Figure 1 shows a typical picture of the secondary X-ray radiation of Co obtained by a scanning electron microscope from the film with $x = 40$. In fact, it can be concluded that the metal macroscopically is distributed uniformly over the area of the sample. And if the metal is localized in the granules, their dimensions are beyond the resolution limit of this technique which is less than 10 nm. So we used indirect method for evaluation of the granules size. It was based on the analysis of magnetization curves considering compositional film as an ensemble of noninteracting superparamagnetic particles of the same size [4]. In our case, the prerequisites for choosing such model were virtually anhysteretic magnetization curves and their adequate approximation using Langevin function. This approach allowed us to obtain the dependence of average grain size d on the composition (for $35 < x < 45$ composition range see figure 1, *b*).

Calculations confirmed granules size being in the nanometer range, and the increase of Co concentration entails significant and almost linear growth of d . Granular structure was directly observed using transmission electron microscopy on a special Co₅₀(Al₂O₃)₅₀ sample, which was deposited onto NaCl crystal and subsequently separated from the substrate (figure 1, *c*). In photography provided numerous dark areas can be identified as Co granules. They are uniform in size (~ 5 nm) and separated by lighter streaks ~ 1 nm thick, which represent the dielectric matrix.

For further experiments on optimization of multilayers properties we have chosen $x = 40$ composition. If we assume that the quantitative results presented above are applicable to ultra-thin composite layers (spacers), the average size of Co grain should exceed 3 nm. In addition, such compositions are characterized by high resistivity values (3.76 Ohm*cm). This suggests good localization of Co granules and can be useful for minimizing the shunting of the responsible for AMR effect permalloy layer.

Figure 2, *a-c* shows examples of hysteresis loops of as-deposited SiO₂/Ta(5)/Fe₂₀Ni₈₀(5)/Fe₅₀Mn₅₀/Fe₂₀Ni₈₀(5)/Co₄₀(Al₂O₃)₆₀(L)/Fe₂₀Ni₈₀(40)/Ta(5) films. They are measured along the axis characterized by a maximum exchange bias. It can be seen that increase of the thickness L of the granular spacer modifies significantly shape of the hysteresis loops. Without spacer two-stage hysteresis loop is observed (figure 2, *a*). It describes the magnetization reversal of both main (in the magnetic field H_{e1}) and auxiliary (in the magnetic field H_{e2}) permalloy layers. Moreover,

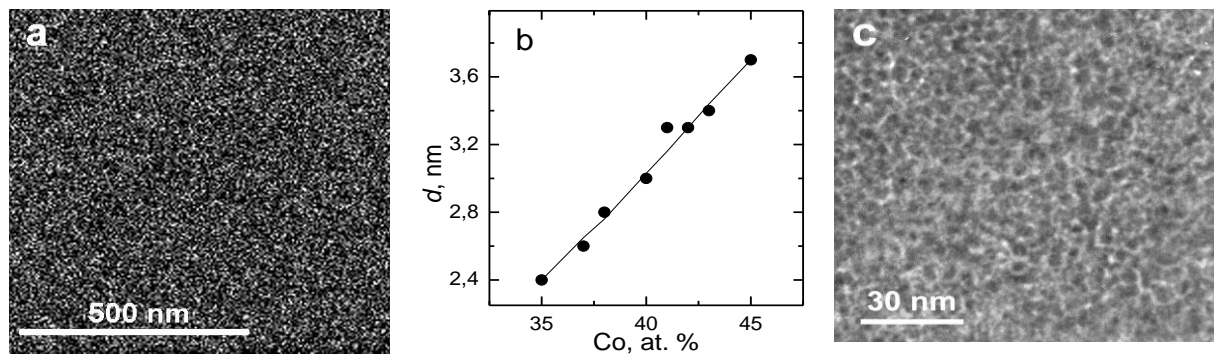


Figure 1. *a* – SEM scan of $\text{Co}_{40}(\text{Al}_2\text{O}_3)_{60}$ film in elemental analysis of Co mode; *b* – dependence of the average grain size on Co concentration; *c* – TEM image of $\text{Co}_{50}(\text{Al}_2\text{O}_3)_{50}$ film obtained by the transmission electron microscope.

both minor loops are shifted along the magnetic field axis which indicates the presence of an interlayer exchange coupling. Introduction of the spacer leads to the three-stage hysteresis loop corresponding to the magnetization reversal of the main permalloy layer and two auxiliary layers of smaller thicknesses (in magnetic fields H_{e2} and H_{e3}). The H_{e1} value of the main layer decreases monotonically with L increasing, and when L reaches 5 nm, it becomes almost zero. At the same time, exchange bias fields of auxiliary layers remain and become almost the same (figure 2, *c*).

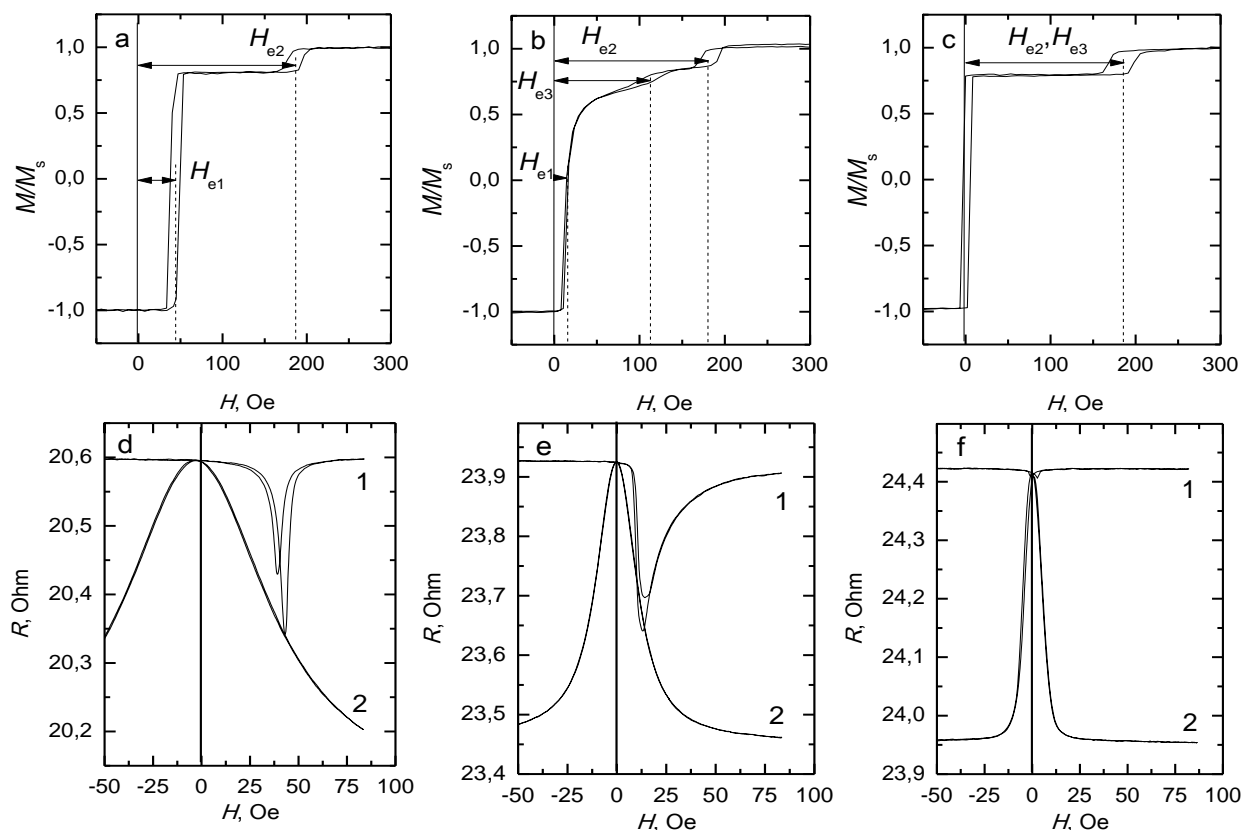


Figure 2. Magnetometric (*a-c*) and magnetoresistive (*d-f*) hysteresis loops measured along (curve 1) and perpendicular (curve 2) to electric current lines on as-deposited $\text{Ta}(5)/\text{Fe}_{20}\text{Ni}_{80}(5)/\text{Fe}_{50}\text{Mn}_{50}(20)/\text{Fe}_{20}\text{Ni}_{80}(5)/\text{Co}_{40}(\text{Al}_2\text{O}_3)_{60}(L)/\text{Fe}_{20}\text{Ni}_{80}(40)/\text{Ta}(5)$ multilayers with different thickness L of the granular spacer: *a, d* – 0 nm; *b, e* – 2 nm; *c, f* – 5 nm.

In general, dependence $H_{e1}(L)$ is shown in figure 3 (curve 1). For comparison, similar $H_{e1}(L)$ dependence obtained for Al_2O_3 spacer is presented (curve 2). It can be seen that in case of the granular spacer the exchange bias field decreases more gradually. This feature can be useful for the intentional tailoring of the exchange bias field. The obvious reason for that is its Co granules, which provide the inter-exchange at relatively high L . The exchange coupling disappearance for $L > 5$ can be associated with excessing of the spacer thickness above the average size of Co grain, which leads to the disappearance of magnetic interlayer bridges. It should be noted, that $H_{e1}(L)$ dependence obtained for as-deposited films and samples subjected to annealing are qualitatively the same. In the last case, however, somewhat higher level of H_{e1} is observed.

Figure 2, *d-f* shows the magnetoresistive loops of as-deposited multilayers with different L . They are measured by application of the external magnetic field along (longitudinal loop) and perpendicular (lateral loop) to the electric current. Probing current in both cases was applied along the induced easy magnetization axis. The amplitude of the applied magnetic field did not exceed H_{e2} and H_{e3} values. Thus, the data obtained reflect hysteresis loops devoted to only main permalloy layer. It can be seen that the longitudinal loops are biased along the magnetic field axes, and the H_e value is close to the value of the exchange bias obtained from magnetometric loops measured on the corresponding samples. Transverse magnetoresistance loops are free from anomalies and have a typical form for magnetouniaxial films. As can be expected, exchange bias is decreasing for large L and resistivity on the transversal loops changes more rapidly. Thus, the introduction of spacer allows to increase sensitivity, but reduces the dynamic range of the medium considered as a material for magnetoresistive sensors. The advantage of the composite spacer is the possibility of a relatively smooth adjustment of these characteristics.

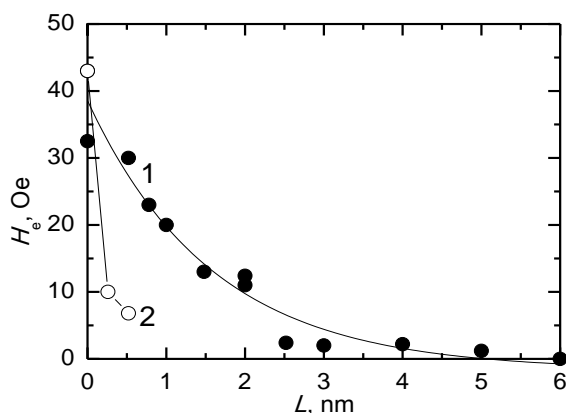


Figure 3. Dependence of the exchange bias field on the thicknesses of $\text{Co}_{40}(\text{Al}_2\text{O}_3)_{60}$ (curve 1) and Al_2O_3 (curve 2) spacers.

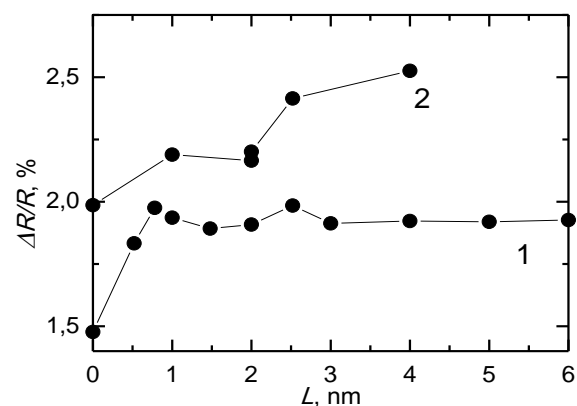


Figure 4. Dependencies of AMR effect on the thickness of the granular spacer of the as-deposited films (curve 1), and annealed films (curve 2).

Figure 4 shows dependences of anisotropic magnetoresistance effect $\Delta R/R$ measured in magnetic field of 60 Oe on thickness of granular spacer on the two types of films. As-deposited samples showed an increase of $\Delta R/R$ with L increasing to ~ 1 nm. For larger thicknesses of spacer magnetoresistance remained almost unchanged and had a value of about 2%. For the annealed films, $\Delta R/R$ grows throughout the considered range and reaches 2.5%. Since measurements of AMR effect were held in the fixed magnetic field, the observed increase of $\Delta R/R$ reflects the weakening of the interlayer coupling and reduction of unidirectional anisotropy caused by introduction of spacers. Annealed films demonstrate stronger exchange interaction, which might lead to the change of $\Delta R/R$ in a larger range of L .

Temperature measurements of the magnetic and magnetoresistive properties were performed on annealed Ta/Fe₂₀Ni₈₀/Fe₅₀Mn₅₀/Fe₂₀Ni₈₀/Co₄₀(Al₂O₃)₆₀/Fe₂₀Ni₈₀/Ta film with $L = 2.5$ nm. Figure 5 shows the temperature dependence of reduced values of the exchange bias field δH_{e1} and the magnetoresistive effect δR . As normalizing values H_{e1} and $\Delta R/R$ at room temperature were used. As can be seen, with temperature increasing in the range from 77 to 320 K, δH_{e1} decreases slightly. The sharp fall of the exchange bias field is observed at higher temperatures. The blocking temperature of this sample was about 400 K. The described behaviour of the exchange bias field is mostly due to the influence of temperature on the magnetic ordering in the antiferromagnetic layer. Besides that transition of Co granules from superparamagnetic to ferromagnetic state can take place with temperature decreasing. The AMR value decreases almost linearly with temperature increasing, which corresponds to the typical temperature behaviour of the anisotropic magnetoresistance effect value due to temperature changes of the mean free path of conduction electrons.

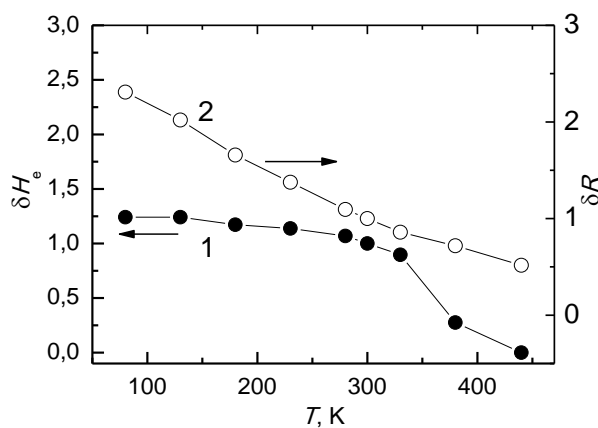


Figure 5. Dependences of reduced values of exchange bias field (curve 1) and the AMR effect (curve 2) on the temperature of the sample with 2.5 nm granular spacer.

4. Conclusion

In the paper we studied the magnetic and magnetoresistive properties of multilayers based on FeMn/Fe₂₀Ni₈₀ with exchange bias, which can be used as a magnetoresistive medium for magnetic field sensors. It was demonstrated that introduction of the composite spacer in the layered structure is an effective way of tailoring its hysteretic properties. The presence of the fine Co granules in the composite allows to extend interlayer interaction for a wide range of the spacer thicknesses. This makes possible the intentional influence on sensitivity of such media. For films with selected layered structure dependences of the exchange bias field and the value of AMR effect on the thickness of the spacer, annealing and temperature are defined.

Acknowledgments

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